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POTENTIAL VISIBLE CHEMICAL LASERS VIA INTERACTIONS OF
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BEERSHEBA (ISRAEL) DEPT OF PHYSI R SALMAN 05 NOV 85
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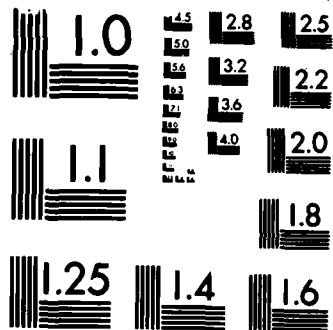
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LARELL K. SMITH, Major, USAF
Chief, Physics/Physical Chemistry



D. L. MOTT
Technical Information Officer

INTRODUCTION

Chemical excitation of metastable electronic states of atoms and small molecules has been suggested as a means of achieving efficient, CW laser operation in the visible (and near IR and UV) wavelength region (see for example, refs. 1 and 2). The advantage of utilizing a metastable state as the upper laser level stems from the fact that for maintaining steady state population inversion, the effective lifetime of the upper level must exceed that of the lower level [1-3]. Favorable lifetime ratios can be obtained in various ways [1,3], but a long radiative lifetime of the upper level eases the burden on the need for fast quenching (or removal) of the lower level. In addition, metastability of the emitting state minimizes the danger of parasitic oscillations (due to high gain). Moreover, long lifetime of the lasing state is essential for using chemical reactions to feed this state since the overall rate of the reactions is limited by the diffusive mixing rate. Indeed, these properties characterize the lasing atom in the first and only electronic transition chemical laser, namely, the CW iodine laser. However, the lasing level is populated by the one step, resonant energy transfer of ~ 1 eV from $O_2(^1\Delta)$ to the I atom and therefore emits in the infrared. The present research is aimed at providing guidelines for obtaining visible chemical lasers via multistep excitation of metastable states by singlet oxygen.

Species with metastable states that might serve as visible (or near visible) CW lasers are heavy atoms with the p^2 , p^3 and p^4 ground-state configuration: Sn and Pb, Sb and Bi, and Se and Te, respectively. Some of the intra-configuration transition probabilities

of these atoms are in the range of $1-100 \text{ sec}^{-1}$ [4] (note that for the lasing transition in the iodine atom the probability is 7.8 sec^{-1}). The energy levels and the strongest transitions within the ground-state configuration of these atoms are presented in table 1. The data is from [4]. Examination of the table reveals that all of the metastable states can be populated as a result of one to three fruitful collisions of a ground state atom with $\text{O}_2(^1\Delta)$ ($E = 7882 \text{ cm}^{-1}$) and/or $\text{O}_2(^1\Sigma)$ ($E = 13121 \text{ cm}^{-1}$) molecule (the latter is obtained from $^1\Delta$ by energy pooling); some of these collisional energy transfer steps are highly resonant.

The reactions of our excited atoms with ground state oxygen have been studied thoroughly (see ref. 5 and refs. cited therein and numerous additional publications by D. Husain and coworkers, mainly in J.C.S. Faraday Trans. II, J. Phys. Chem. and J. Photochem. from 1974 on), but very little is known about the total cross section for reactions of singlet oxygen with these atoms or about the branching ratios of chemical reactions vs. energy transfer. It is expected that when the chemical reaction is endoergic or only slightly exoergic, energy transfer will prevail. Fortunately, the reactions of ground state O_2 with our atoms, except Sn, are very endoergic ($> 0.7 \text{ eV}$) [6]; the reactions of $^1\Delta$ are either endoergic (with Pb, Bi and Te) or slightly exoergic (with Sb and Se) and the reaction of $^1\Sigma$ is thermoneutral with Bi and slightly exoergic with Pb and Te. It is expected, therefore, that in a fast flow system, these reactions will result, mainly, in atomic excitation in the region closest to the O_2 /metal atoms mixing zone. Further downstream

Table 1. Energy levels (observed, cm^{-1}) and strongest transition probabilities (calculated, sec^{-1}) for p^2 , p^3 and p^4 configurations. The data is from [4].

A. p^2 configurations

Level	SnI($5p^2$)	PbI($6p^2$)	Transition ^a	Type	SnI	PbI
$3P_0$	0	0	$1S_0 - 3P_2$	EQ ^b		10
$3P_1$	1692	7819	$1S_0 - 3P_1$	MD ^c	7.0	78
$3P_2$	3428	10650	$1D_2 - 3P_2$	MD		12
$1D_2$	8613	21458	$1D_2 - 3P_1$	MD		14
$1S_0$	17163	29467	$3P_1 - 3P_0$	MD		7.3

B. p^3 configurations

Level	SbI($5p^3$)	BiI($6p^3$)	Transition	Type	SbI	BiI
$4S_{3/2}$	0	0	$2P_{3/2} - 2P_{1/2}$	MD		8.6
$2D_{3/2}$	8512	11419	$2P_{3/2} - 2D_{5/2}$	MD	1.5	23
$2D_{5/2}$	9854	15438	$2P_{3/2} - 2D_{5/2}$	EQ		10
$2P_{1/2}$	16396	21661	$2P_{3/2} - 2D_{3/2}$	MD	4.0	120
$2P_{3/2}$	18465	33165	$2P_{3/2} - 2D_{3/2}$	EQ		4.5
			$2P_{1/2} - 2D_{3/2}$	MD	1.1	1.2
			$2P_{3/2} - 4S_{3/2}$	MD	5.2	7.3
			$2P_{1/2} - 4S_{3/2}$	MD	3.3	55
			$2P_{1/2} - 4S_{3/2}$	EQ		6.2
			$2D_{5/2} - 4S_{3/2}$	MD		6.4
			$2D_{3/2} - 4S_{3/2}$	MD	1.1	31

C. p^4 configurations

Level	SeI($4p^4$)	TeI($5p^4$)	Transition	Type	SeI	TeI
$3P_2$	0	0	$1S_0 - 1D_2$	EQ	2.3	3.1
$3P_1$	1989	4751	$1S_0 - 3P_1$	MD	7.7	37
$3P_0$	2534	4707	$1D_2 - 3P_2$	MD		3.3
$1D_2$	9576	10559	$3P_1 - 3P_2$	MD		2.2
$1S_0$	22446	23199				

^a Only transitions stronger than 1 sec^{-1} are listed; ^b Electric quadrupole; ^c Magnetic dipole.

the excited atoms may react chemically with O_2 and their reactions can be studied. It should be noted that these reactions produce metal monoxides. Examination of the energy levels of the monoxides of our atoms [6], reveal that they can be excited by energy transfer from singlet oxygen.

The present research has been the first part of a continued effort to thoroughly study the kinetics and spectroscopy of the heavy- atoms/singlet oxygen mixtures. The measurements were conducted in a flowing afterglow reactor to which a metal evaporation furnace and a singlet oxygen generator are interfaced [7]. The atoms studied so far were Pb and Bi.

EXPERIMENTAL

1. New design of the reactor.

In the reactor used in our preliminary studies of metal-atoms/singlet oxygen reactions [7], clogging of the furnace precluded its prolonged (for more than about an hour) and stable operation, especially at high temperatures. Therefore, a new design of the reactor was tried [8]. The main reason for clogging in the previous reactor was the reaction of oxygen with metal vapor at the outlet of the furnace. In the new reactor the oxygen was introduced 14 cm downstream of the furnace outlet and, thus, clogging problems were completely eliminated.

Another problem encountered in the previous reactor was coating of the observation windows by metal and metal oxide vapors. Purging the windows with inert gas, using a narrow hose aimed at the center of the window, reduced but did not eliminate coating. In the new reactor a symmetric, homogenous and fast flow of gas from the circumference toward the center of the window was applied. With this method of purging, about 60 hours of reactor operation (as compared to a couple of hours in the previous reactor) was possible before coating on the window was detectable.

A useful change in the reactor, that will be fully exploited in future experiments, was the addition of observation windows to the flow tube downstream to the metal-atoms/oxygen mixing zone to allow monitoring of the reactive species at various observation points.

2. Singlet oxygen generation

In the present experiments the singlet oxygen was produced by operating microwave discharge in oxygen (see ref. 7 for details). Using this method, $O_2(^1\Delta)$ in concentrations of up to ~ 10% can be obtained [9]. It is planned that in future experiments a chemical generator, capable of producing higher concentration of $O_2(^1\Delta)$, will be used. This generator has already been built. It is similar to the one described in ref. 10 except that the Cl_2/H_2O_2 reactor where $O_2(^1\Delta)$ is produced is spherical rather than cylindrical (in order to minimize splashing when large amounts of reagents are used) and that a closed cycle cooling system for the 195 K water vapor trap (see ref. 10) was added.

3. Detection of reaction products

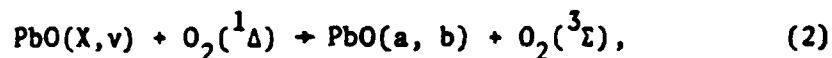
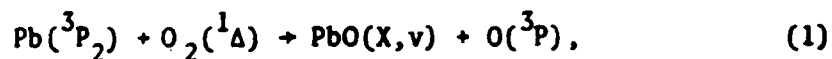
The species produced via the metal-atom/singlet oxygen interaction were monitored using laser induced fluorescence (LIF) and emission spectroscopy techniques. For the LIF measurements, an excimer pumped dye laser (Lambda Physik EMG101MSC/FL2002) was used. The laser beam was shined along the center of the flow tube and the induced fluorescence was monitored at right angle to the beam through the observation windows via a filter/photomultiplier/amplifier/recorder system.

For luminescence measurements a monochromator/photomultiplier/picoammeter/recorder system was used. The monochromator (0.5 m Spex model 1870) was equipped with grating blazed for 500 nm (first order). The photomultiplier tube (Hamamatsu R-928) was thermoelectrically cooled.

RESULTS AND DISCUSSION

1. LIF measurements in the Pb/O₂ mixture

From our preliminary experiments conducted in the Pb/O₂ system it was concluded that electronically excited PbO is produced via the reaction sequence [7]



where a, b denote the lowest electronically excited states of PbO (see fig. 1). It was argued in ref. 7 that since the a and b states

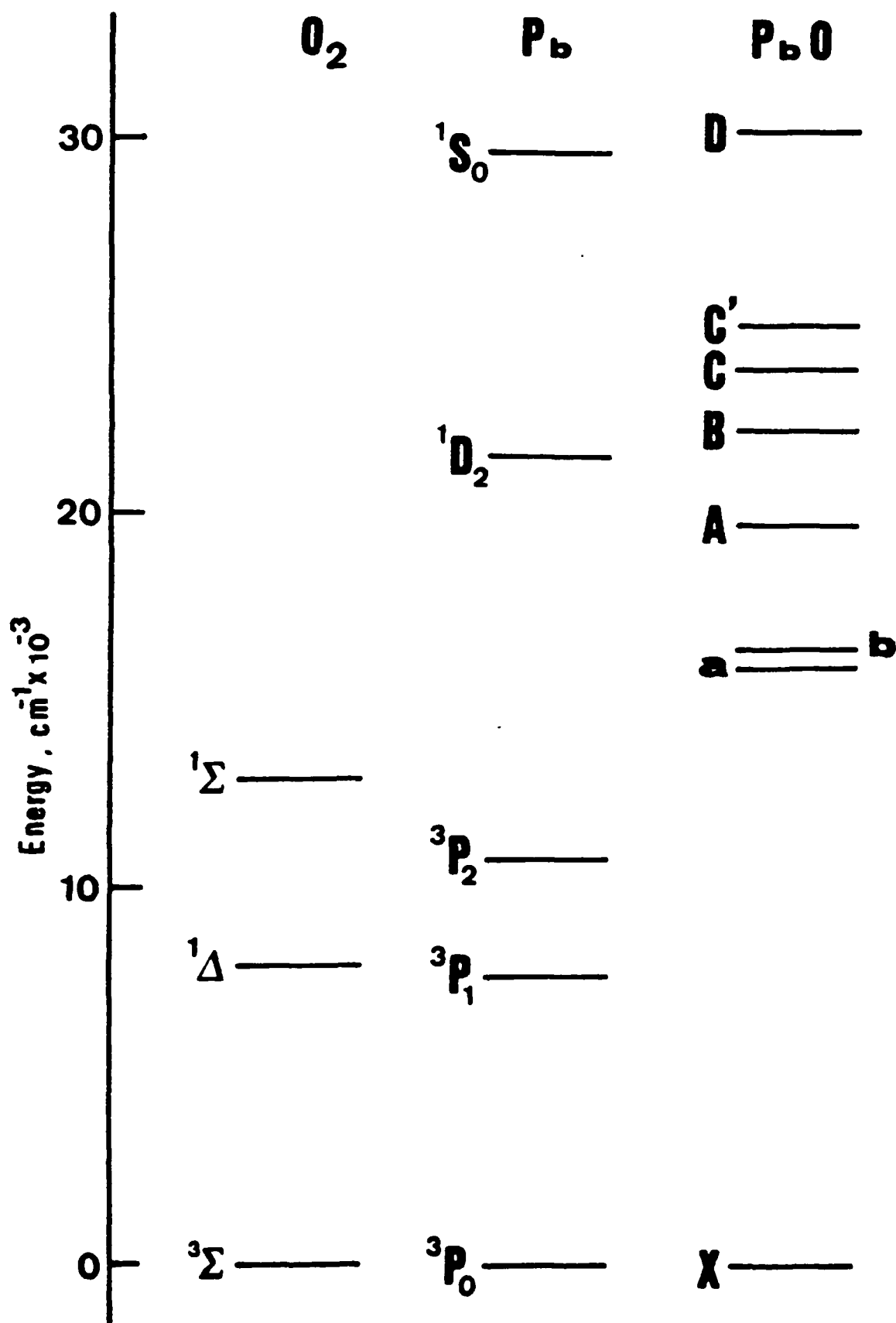


Figure 1. The lowest energy levels of O_2 , Pb and PbO .

lie $\approx 2\text{eV}$ above $\text{PbO}(X, v = 0)$ whereas energy transfer from $\text{O}_2(^1\Delta, ^1\Sigma)$ can provide $\approx 1.6\text{eV}$, the PbO precursor must be vibrationally excited to $\approx 0.4\text{eV}$. Our first application of the LIF technique in the present research has been to monitor the vibrational population of $\text{PbO}(X)$ in order to establish the excitation mechanism.

PbO transitions were excited at wavelengths around 450 nm using coumarin dyes. Vibrational population of $\text{PbO}(X)$ up to $v = 5$ was easily detected. These LIF measurements thus corroborate the above mentioned excitation mechanisms (note that $v = 5$ lie $\sim 0.4\text{eV}$ above $v = 0$). Further spectroscopic studies of the PbO states produced in the Pb/O_2 mixture are now in progress and will be reported in the future.

2. Emission from the Bi/O_2 mixture

Mixing of ground state oxygen with bismuth vapor did not result in any visible reaction or measurable amount of excited species. When the microwave discharge was operated on the O_2 a reddish, diffuse flame was produced, extending to about 10 cm downstream of the mixing zone, and emission from excited O_2 , Bi , Bi_2 and BiO was observed. The lowest energy levels of O_2 , Bi , Bi_2 and BiO are presented in fig. 2. The reddish color changed to whitish upon increasing the amount of O_2 and many unassigned bands appeared in the spectrum, probably due to emission from polyatomic oxides of Bi . The results reported herein were obtained from the reddish flame; the O_2 pressure was from $0.1 - 0.3$ Torr and that of the Ar carrier gas for the Bi vapor 2.2 Torr,

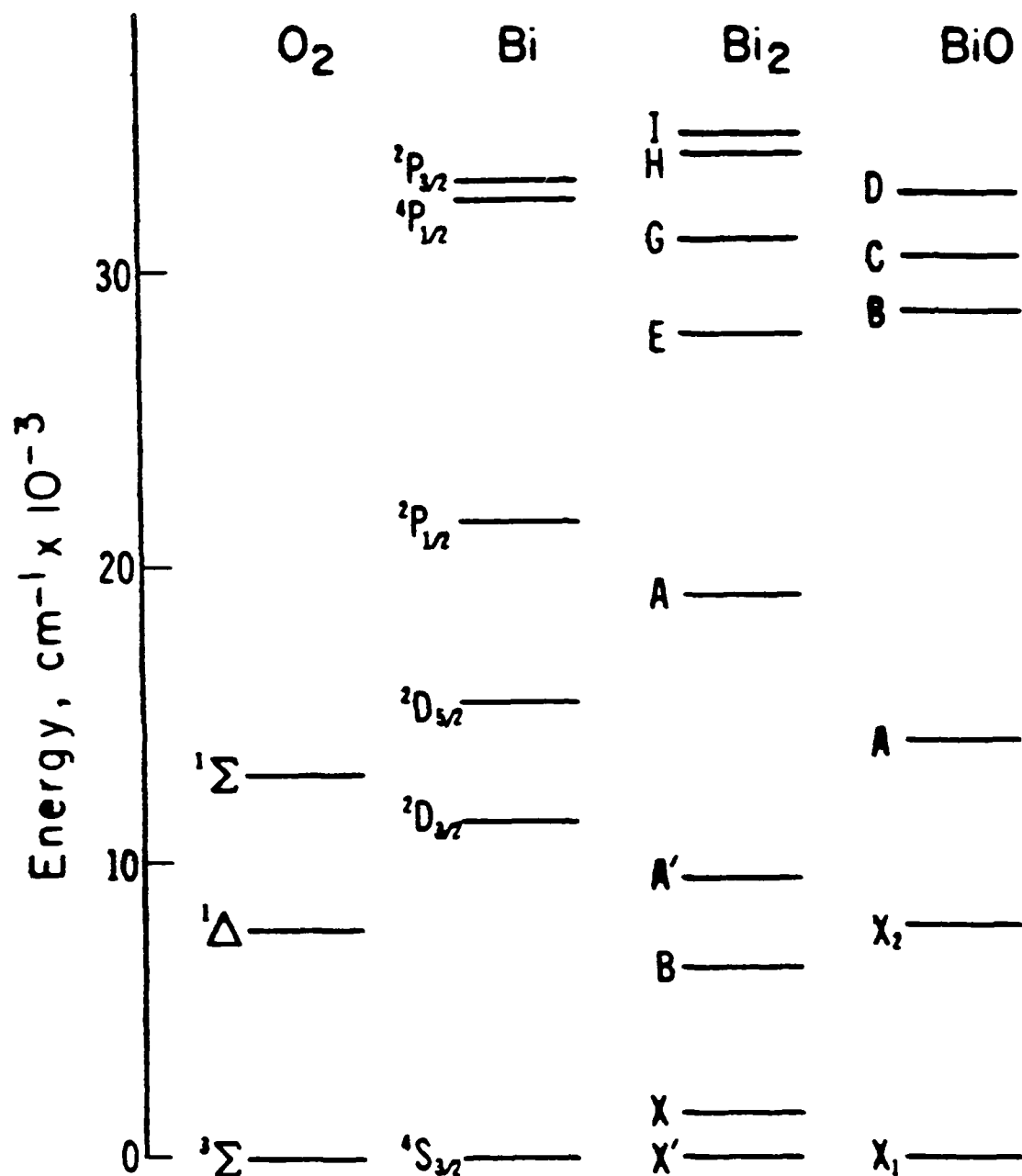


Figure 2. The lowest energy levels of O_2 , Bi , Bi_2 and BiO .

the Ar purge gas added 0.1 Torr to the total pressure (2.4 - 2.7 Torr). The concentration of bismuth was not measured, but from comparison to measurements of copper vapor concentration carried out using the same furnace [11] we estimate that the former is limited within the range of $10^8 - 10^{13}$ atom cm^{-3} at the observation window for crucible temperature from 400 - 800°C. Note that the bismuth vapor consists of ~ 50 - 70 % Bi_2 molecules in this temperature range, the rest being Bi atoms [12,13].

The emission spectrum from 300 - 900 nm was recorded as a function of the crucible temperature at fixed flow conditions. The following transitions have been observed.

- a) O_2 : $^1\Sigma_g^+, v = 0 \rightarrow ^3\Sigma_g^-, v = 0$ at 762 nm.
- b) Bi : $^2D_{3/2} \rightarrow ^4S_{3/2}$ at 875 nm.
- c) Bi_2 : $A(^0\text{u}, v = 0-15) \rightarrow X(^1\Sigma_g^+, v = 0-12)$ from 550-640 nm.
- d) BiO : $A(^2\Pi_{1/2}, v = 1-24) \rightarrow X_1(^2\Pi_{1/2}, v =)-11)$ from 550-800 nm.

Detailed analysis of the spectra is now in progress [14]. However, from the general features of the spectra and from the temperature dependence of the intensities of the emission from the excited species, presented in table 2, some interesting conclusions regarding the kinetics of the Bi/O_2 system can be drawn.

The most striking features of the spectra are the intensity of the forbidden $\text{Bi}(^2D_{3/2} - ^4S_{3/2})$ line at 875 nm at crucible temperatures

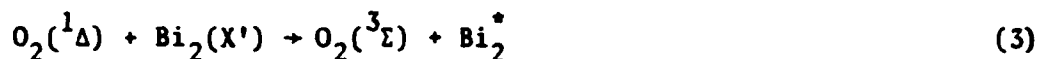
Table 2. Emission intensities from excited species is the Bi/O₂ mixture (O₂=0.1, Ar=2.3 Torr).

Intensities are peak heights, in arbitrary units, of the listed transitions as reduced from the spectrum. The spectrum was not corrected for the instrument wavelength response.

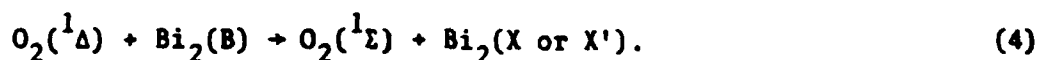
Crucible temperature (°C)	O ₂ (¹ Σ,0→ ³ Σ,0)	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})	Bi(² D _{3/2} → ⁴ S _{3/2})
	762 nm	875 nm	600 nm	588 nm	594 nm	593 nm			
400	2	0	0	0	0	0			
450	3	2	4	4	4	3			
500	4	4	11	17	16	9			
550	16	66	60	102	138	45			
600	227	877	346	410	142	91			
650	70	1408	422	666	544	269			
700	20	390	56	91	77	56			
750	4	25	7	11	12	7			
800	0	0	0	0	0	0			

a) The temperature dependence of the intensities was similar for all vibronic bands belonging to this transition.

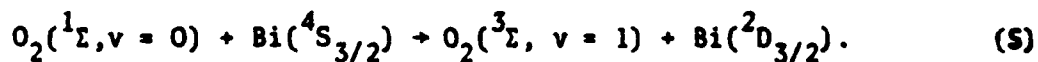
around 650°C (note that the instrument wavelength response was relatively poor at this wavelength) and the increase of the $O_2(^1\Sigma)$ emission intensity with temperature (i.e. with Bi and Bi_2 concentration) up to 600°C. Since the only excited species initially available in significant concentrations in the reaction mixture is $O_2(^1\Delta)$ (note that $\frac{[{}^1\Sigma]}{[{}^1\Delta]}$ downstream of the discharged $O_2 \lesssim 10^{-3}$ [15]), the first step of excitation of other species must involve $O_2(^1\Delta)$. Since excitation/reaction of ground state Bi by/with $O_2(^1\Delta)$ is energetically excluded (note also that four center reaction of Bi_2 with O_2 to produce $2BiO$ is very improbable), the initial excitation step in the reaction mixture must be



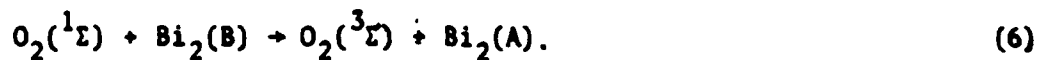
where the asterik denotes excitation to either the X or the B state (see fig. 2). The increase of $O_2(^1\Sigma)$ concentration with that of Bi_2 is most probably due to the energy pooling process



Enhancement of $O_2(^1\Sigma)$ emission due to similar energy pooling processes have been observed in O_2/I_2 [16] and O_2/Pb mixtures [7]. $Bi(^2D_{3/2})$ can then be formed via the spin-allowed, resonant energy transfer process



$Bi_2(A)$ can similarly be produced:



Once the metastable $\text{Bi}(^2\text{D}_{3/2})$ and $\text{O}_2(^1\Sigma)$ have been produced in significant amounts, several channels are open for reaction of Bi with O_2 (where at least one of them is excited to $\geq 1.6\text{eV}$) to produce BiO . Excitation of BiO to the A state can then take place via collisions with $\text{O}_2(^1\Delta$ or $^1\Sigma)$ (see fig. 2). Note that the short lived $\text{Bi}_2(\text{A})$, with radiative lifetime of a few hundreds of nanoseconds [13], is not expected to react significantly under the present experimental conditions (relatively low concentration of potential reactive partners).

The reaction scheme outlined above accounts for the simultaneous production of excited species observed in the reaction mixture. It also is consistent with the fact that upon increasing the temperature from 600 to 650°C , $\text{O}_2(^1\Sigma)$ emission intensity decreases whereas that of the other species increases. Probably, when $[\text{Bi}]$ and $[\text{Bi}_2]$ rise above certain limit the consumption rate of $\text{O}_2(^1\Sigma)$ in reactions (5) and (6) and in reactions producing BiO surpasses its production rate in reaction (4). At still higher concentration of Bi and Bi_2 , most $\text{O}_2(^1\Delta)$ is consumed in reaction (3) and production of all emitting species is suppressed (see table 2).

SUMMARY

The results of the measurements carried out in Pb/O_2 and Bi/O_2 mixtures show that significant electronic excitation of metal atoms, metal dimers and metal oxides can be obtained via multiple collisions with singlet molecular oxygen. The mechanisms for Pb^* and PbO^* production have previously been explained on the basis of emission and absorption spectroscopy

studies [7]. The present LIF measurements corroborate the mechanism for PbO^* formation.

Mechanisms for production of excited species in Bi/O_2 are inferred from the dependence of the emission intensity from these species on $[\text{Bi}]$ and $[\text{Bi}_2]$. In particular, it is suggested that the strong emission from $\text{Bi}(^2\text{D}_{3/2})$ must originate from energy transfer from $\text{O}_2(^1\Sigma)$ which, in turn, is produced via multiple collisions of $\text{O}_2(^1\Delta)$ with Bi_2 .

The results of the present research are encouraging regarding the prospects of obtaining significant concentrations of atomic and molecular species via multiple collision with singlet molecular oxygen. For assessing the potential of this method for achieving short wavelength chemical lasers, more spectroscopic and kinetic work is needed. Some work on this subject is now in progress in our laboratory and will be published in the near future.

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